Optical multistability and light self-modulation at double resonance

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It is shown that in a system of atoms or molecules, an instability arises in a resonator at double optical resonance conditions, because of the high-frequency Stark effect, caused by the collective field, and this leads to a number of new cooperative threshold optical phenomena.

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The bistable behavior, recently found in a number of nonlinear-optical systems,[1–8] is attracting a great deal of interest as an example of a kinetic phase transition having important technical applications (bistable optical memory units, etc.). This paper reports on new cooperative threshold optical phenomena, appearing at double resonance conditions, which may be of interest for building multifunctional optical elements with adjustable parameters.

Let us consider a system of three-level atoms or molecules ($\epsilon_1 < \epsilon_2 < \epsilon_3$) located in an optical cavity, tuned to a frequency $\sim\omega_{32}$ [$\omega_{ij} = (\epsilon_i - \epsilon_j)/h$]. Radiation E_i having a frequency $\Omega = \omega_{32}$ is introduced into the cavity, and narrow-band optical pumping E_p with a frequency $\Omega_p = \omega_{21} + \Delta$ ($\Delta \ll \omega_{ij}$ is the resonance detuning) is performed at the 1–2 transition. We shall assume that the cavity exerts no influence on the pumping radiation (for example, it is done at an angle to the optical axis of the cavity or the mirrors are transparent in the appropriate frequency region). The intracavity field E of frequency Ω depends both on E_i and on the polarizability of the medium at this frequency. Since levels 2, 3 are not populated in the absence of pumping, the basic, resonance part of this polarizability is determined by the absorption of the pumping. In turn, due to the action of the field E, the pumping absorption band is split into two Stark components, separated by $\sim 2\Omega_R$ ($\Omega_R = |d_{32}E|/h, d_{ij}$ are the dipole matrix elements). In this situation the absorption of the pumping at the fixed frequency Ω_p changes and the resulting feedback leads, under certain conditions, to an instability in the system. For a quantitative description let us consider a high-Q cavity, the entrance and exit mirrors of which have an intensity transmission coefficient $T \ll 1$, and the others are perfectly reflecting. If the cavity is not too long and the density N of atoms not too large, then the "average field" approximation is applicable, which is widely used to describe the optical bistability phenomenon in two-level absorbers[2,5,6] and its two-photon analogs.[7,8] The modulus $\mathscr{E}$ and phase ψ of the average field $E = \mathscr{E} e^{i\psi}$ satisfy the equations

$$\frac{d\mathscr{E}}{d\tau} = -\mathscr{E}\left[1 + \frac{2\pi NkL}{T}\chi''(\mathscr{E})\right] + \frac{E_i}{\sqrt{T}}\cos\psi, \tag{1}$$

$$\mathscr{E}\frac{d\psi}{d\tau}=\mathscr{E}\left[\phi+\frac{2\pi NkL}{T}\chi'(\mathscr{E})\right]-\frac{E_i}{\sqrt{T}}\sin\psi,$$

$$\tau=\mathscr{Z}/cT,\quad \phi=\theta/T$$

Here $\chi(\mathscr{E})=\chi'+i\chi''$ is the atomic polarizability at frequency Ω, $k=\Omega/c$, c is the velocity of light, L is the length of the sample with the substance; $\mathscr{L}$ is the total resonator length, θ is its detuning ($k\mathscr{L}=2\pi m+\theta$, m is an integer, $\theta \ll 1$). The equation for the steady-state value of $\mathscr{E}$ has the form

$$Q(\mathscr{E})-\frac{E_i^2}{T}=0,\quad Q(\mathscr{E})=\mathscr{E}^2\left\{\left[1+\frac{2\pi NkL}{T}\chi''(\mathscr{E})\right]^2+\left[\phi+\frac{2\pi NkL}{T}\chi'(\mathscr{E})\right]^2\right\}. \tag{2}$$

The following conditions must be satisfied for the stability of the steady-state solutions of (1):

$$\frac{dQ}{d\mathscr{E}}>0,\quad \frac{dP}{d\mathscr{E}}>0,\quad P(\mathscr{E})=\mathscr{E}^2\left[1+\frac{2\pi NkL}{T}\chi''(\mathscr{E})\right]. \tag{3}$$

Since the general expression for $\chi(\mathscr{E})$ is cumbersome, let us consider the cases $|\Delta|\gg\gamma$ and $|\Delta|\ll\gamma$ separately; where γ is of the order of the widths of the lines ω_{31}, ω_{21}. *The* $|\Delta|\gg\gamma$ *case*. Dispersion effects play the predominant role for these conditions. In dimensionless variables Eq. (2) has the form ($\Delta>0$):

$$y_1^2=x_1^2\left\{1+\left[\phi-\frac{2C_1}{(x_1-1)^2+\delta^2}\right]^2\right\},$$

$$x_1=\frac{1}{\sqrt{T}}\frac{|d_{32}|E_t}{\hbar\Delta},\quad y_1=\frac{1}{\sqrt{T}}\frac{|d_{32}|E_i}{\hbar\Delta},\quad \delta=\gamma/\Delta, \tag{4}$$

$$C_1=\frac{1}{2}\frac{\pi NkL}{T}\frac{\gamma}{\Delta^3}\frac{|d_{32}|^2}{\gamma_{32}}\frac{|d_{12}|^2E_p^2}{\hbar^2}.$$

Here $E_t=\sqrt{T}\mathscr{E}$ is the amplitude of the light leaving the resonator, γ_{32}, $\gamma_{31}=\gamma_{21}=\gamma$ are the transverse relaxation rates of the corresponding atomic transitions. When the pumping intensity exceeds certain critical values, Eq. (4) can have two and, unlike Refs. 1–3, 5–8, even three stable solutions. The last is due to the fact that because of

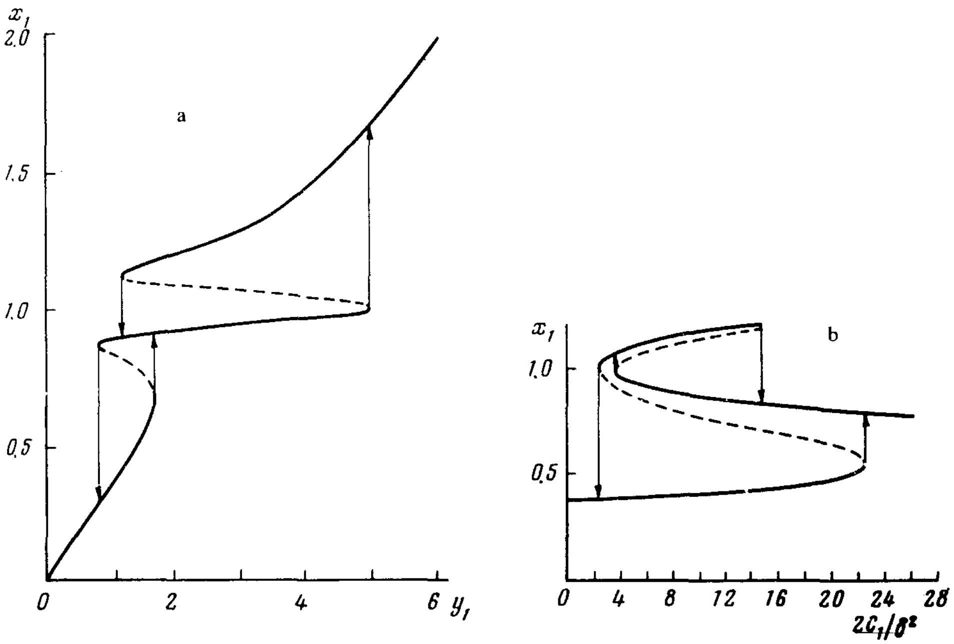

FIG. 1. Hysteretic dependences of the amplitude of the light leaving the cavity on the following: a) $y_1,(2C_1/\delta^2 = 8)$, b) $C_1,(y_1, = 1.2)$. $\delta = 0.1$, $\phi = 3$.

our nonmonotonic $\chi'(\mathscr{E})$ dependence the system, in contrast to Refs. 3, 6, can be found in resonance twice with the same resonator mode as $\mathscr{E}$ increases continuously. Examples of the multivalued dependences of the amplitude of the light, passing through the resonator, on y_1 (for $C_1 =$ const) and C_1 (for $y_1 =$ const) are shown in Fig. 1,a,b. The unstable branches are shown dashed, and the arrows indicate the abrupt changes in E_t. The hysteretic character of the curves means the presence of an "optical memory" in the system. In this case, unlike the case in Refs. 1–7, the cooperativeness parameter C_1 and the "switching" threshold depend on the pumping intensity and are adjustable.

The $|\Delta| \ll \gamma$ case. In this case the absorption properties of the medium play the principal role ($\chi'/\chi'' \ll 1$). The "equation of state" (2) has the form

$$y_2^2 = x_2^2\left\{\left[1 + \frac{2C_2}{(1 + x_2^2)(1 + \beta x_2^2)}\right]^2 + \phi^2\right\}, \tag{5}$$

$$x_2 = \frac{1}{\sqrt{T}}\frac{E_t}{E_s}, \quad y_2 = \frac{1}{\sqrt{T}}\frac{E_i}{E_s}, \quad E_s = \frac{\hbar}{|d_{32}|}\left[\frac{\gamma_{32}\,w_{21}}{2}\,\frac{w_{31} + w_{32}}{w_{31} + w_{21}}\right]^{1/2},$$

$$C_2 = \frac{\pi NkL}{T}\left(\frac{2\gamma}{w_{21}} + 1\right)\frac{|d_{32}|^2}{\hbar\gamma_{32}}\,\frac{|d_{12}|^2 E_p^2}{\hbar^2\gamma^2}, \quad \beta = \frac{w_{21}\,\gamma_{32}}{2\gamma^2}\,\frac{w_{31} + w_{32}}{w_{31} + w_{21}},$$

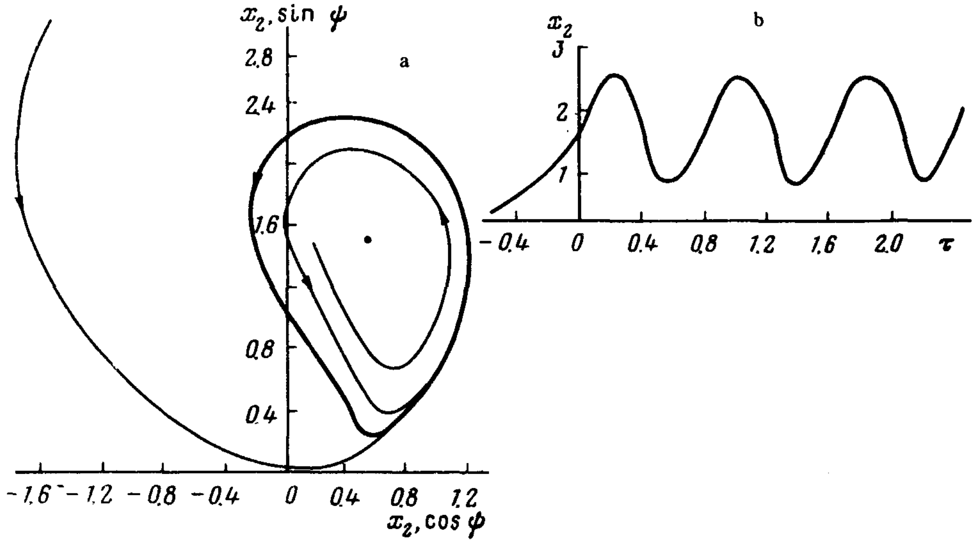

FIG. 2. a—Limiting cycle in phase plane of system. The unstable equilibrium condition is denoted by point; thin lines are examples of phase trajectories. $\phi = 10, C_2 = 20, y_2 = 17$; b—corresponding time dependence of the amplitude of light leaving cavity.

where E_s is the saturation field of the 2–3 transition, w_{ij} are the longitudinal relaxation rates of the corresponding transitions. When the pumping exceeds some threshold in the system, a bistable regime can also be realized with adjustable switching thresholds. The most significant feature, however, distinguishing this case from absorption optical bistability, due to the saturation effect in two-level absorbers, is the possibility of a nonmonotonic dependence of the power $\sim P(\mathscr{E})$ on the field E at sufficiently large C_2. Under certain conditions this leads to the fact that none of the steady-state solutions of the system is stable. In this situation a limiting cycle appears in the phase plane of the system of equations, and an amplitude-phase self-modulation of the light leaving the cavity arises at fixed intensities of the excitation fields (see Fig. 2,a,b). The resulting time ordering can be placed in the class of transient dissipative structures that appear in a number of highly nonresonant open systems.[9]

The discussed effects can be observed both in solid-state samples, where the necessary three-level system is present, for example in the impurity absorption region, and also in gas-filled cells. Estimates show that the requirements imposed for this are essentially no more stringent than those necessary for observing optical bistability in cavities with two-level absorbers.[1–3,5–8]

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